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PATENT SPECIFICATION

DRAWINGS ATTACHED

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COMPLETE SPECIFICATION

Improvements in Autoclaves

We, CIBA LIMITED, a body corporate organised according to the laws of Switzerland, of Basle, Switzerland, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly decribed in and by the following statement:—

This invention provides an improved autoclave having a high capacity for thermal loading, that is to say, an autoclave for carrying out processes which require a high throughput of heat.

It is often necessary in the chemical industry to carry out batchwise reactions under relatively high pressures, and for this purpose autoclaves are used. The greater the pressure and the greater the volume of the autoclave the thicker must be the wall of the vessel in order to withstand the pressure in the interior of the autoclave. For this reason the dimensions of an autoclave are limited due to the necessity to keep the dimensions relatively small in order to avoid serious disadvantages. On the other hand it is often desirable in the interests of economy that the autoclave should have as large a volumetric capacity as possible.

The difficulties referred to above become even more acute when processes requiring a high throughput of heat are carried out, that is to say, when it is necessary that the autoclave should have a high capacity for thermal loading. By capacity for thermal loading is meant the ability of the autoclave to receive and to have withdrawn therefrom relatively large amounts of heat in a short time. The capacity for thermal loading may also be called thermal manoeuvrability. This requirement exists principally when it is desired to carry out reactions which are either strongly exothermic or strongly endothermic. However, an autoclave having a high capacity for thermal loading is also necessary for other reactions, which have to be carried out at very high or very low temperatures, because only then can the period required for heating up or cooling the autoclave be kept short. Long periods for heating up or cooling are disadvantageous for productivity, because during these periods the autoclave is not being utilised for carrying out the reaction.

It is obvious that the need for an autoclave of large volumetric capacity to have thick walls interposes a relatively large resistance to the transfer of heat between the exterior of the autoclave, which is to be cooled or heated externally, and the material undergoing reaction. Furthermore, the problem is rendered more acute by the fact that the rapid cooling or heating of such an autoclave gives rise to stresses in the vessel wall, which are superposed in an unfavourable manner upon the tensional stresses due to the internal pressure. Thus, heat stresses arise which become manifest in the material of the reaction vessel as tensional stresses on the inner side during heating up and on the outer side during cooling. As, in addition, the reaction vessel is strongly stressed by the increasing pressure, it is apparent that such an autoclave has only a low capacity for thermal loading under the full working pressure. Accordingly, the necessary high capacity for thermal loading can in some cases only be achieved with an autoclave of relatively small dimensions.

An autoclave can be given a high capacity for thermal loading by bringing about the heat exchange, not through the wall of the autoclave, but by means of a coil disposed in the interior of the autoclave. The coil may serve either for heating or cooling, and the surfaces of the coil available for heat exchange can be generously dimensioned without difficulty. However, this solution of the problem does not apply to numerous reactions, in which it is necessary to provide a good stirring action extending to the whole contents of the vessel, especially when the reaction mixture is relatively viscous or becomes viscous during the reaction, or when appreciable quantities of solid substances are present or formed during the reaction. Accordingly, an autoclave which has a substantially smooth inner wall and contains no heating or cooling coil would be preferable. In many cases autoclaves having cool-

ing or heating coils disposed therein are totally unsuitable.

Autoclaves have been proposed in which a pressure-resistant outer casing is provided within which is located the reaction vessel itself, and in which a space between the casing and the reaction vessel is filled with a medium, such as a liquid or gas, that transmits the pressure within the reaction vessel to the casing, so that the reaction vessel need only have a thin wall. It has also been proposed to use the pressure medium in the intervening space also for heating or cooling the reaction vessel. In this case, however, a very large amount of the heat to be supplied or withdrawn reaches the outer casing, so that a considerable temperature gradient exists in the wall of the autoclave.

The present invention provides an autoclave for carrying out processes operated at high throughputs of heat, which comprises an outer casing capable of withstanding the full working pressure within the autoclave, a relatively thin-walled inner reaction vessel, heat exchange forming part of, or in thermal contact with the wall of the reaction vessel for supplying heat energy to or withdrawing it from the said vessel, heat-insulating means interposed between the reaction vessel and the pressure-resistant outer casing, and means for maintaining in the space between the reaction vessel and the outer casing a pressure approximately equal to the pressure in the reaction vessel.

Apparatus constructed in accordance with the invention is illustrated by way of example in the accompanying drawings in which:

Fig. 1 shows in vertical cross-section an autoclave having an interior reaction vessel,

Figs. 2, 4 and 9 show other forms of such autoclaves, and

Figs. 3, 5, 6, 7 and 8 show constructional details of the various forms of autoclaves.

Fig. 1 shows an autoclave of which the pressure-resistant outer casing consists of a lower part 1 and an upper part 2. Within the exterior casing is an interior reaction vessel which has relatively thin walls and consists of a lower part 3 and an upper part 4. The lower part 3 is provided on its outer side with a welded pressure-resistant coil 5, for example, having a semi-tubular cross-section. The space between the exterior casing and the interior reaction vessel is rendered heat-insulating in accordance with the invention, for example, by means of a filling of a ceramic heat-insulating material. Advantageously the space between the exterior casing and the interior reaction vessel is connected through an automatic pressure equalising device with a source of compressed gas in such manner that the pressure in the said space is maintained approximately equal to the pressure inside the reaction vessel.

The exterior pressure-resistant casing may be constructed in a manner similar to that of known autoclaves. Since this casing does not come into contact with the substances taking part in the chemical reaction, it is advantageously constructed of a material well adapted to take a high pressure without having to take into account the chemical resistance of the material used for this purpose. Thus, it may be made, for example, of cast steel. The insulating material between the exterior casing of the reaction vessel must be so constructed that it can transmit to the exterior casing the pressure prevailing in the reaction vessel without deformation of the latter, or that, in the case of the pressure equalisation described below, it offers no resistance to equalisation of the gas pressure within the said space. Advantageously, there is used an insulation consisting of Raschig rings, Berl saddle-shaped bodies, stoneware balls or the like, or a heat insulating fluid.

It is of advantage that the heat exchange means associated with the interior reaction vesel should be thermally connected to the vessel, for example in the form of a coil for example, of semi-tubular cross-section, welded to to the outer side of the vessel. The connections for the heating or cooling medium to be circulated in the heat exchange means may be passed through the exterior casing in any desired manner, advantageously with the interposition of a certain amount of heat insulating material. If the heat transfer coil is made pressure-resistant the heating or cooling medium may be supplied from the exterior under atmospheric pressure.

In operating the autoclave it is of advantage to maintain in the space between the exterior casing and the interior reaction vessel a certain pressure, which is hereinafter referred to as the equalisation pressure, and which is derived from a source of gas pressure independent of the chemical reaction. In the simplest case the equalisation pressure may be derived from one or more bottles of compressed gas. In many cases it is desirable to use for this purpose an inert gas, for example, nitrogen. The gas used for pressure equalisation has the important function of relieving the interior reaction vessel from the high reaction pressure, which the vessel cannot withstand owing to its thin walls. On the other hand, owing to its thin walls the reaction vessel is an efficient heat exchanger, since thermal stresses therein are of a low order due to the thin walls. As shown in Fig. 1 the pressure in the space between the reaction vessel exterior casing is supplied through the conduit 6 to the control system. Pressure equalisation acts on the control means through the conduit 7. The conduit 7 also serves for the supply and withdrawal of the inert gas, for example, nitrogen.

The control system, as shown in the drawing, consists of two pressure regulators 8 and 9. The pressure regulator 8 is connected to a

pressure bottle 10, which contains the inert gas, advantageously nitrogen. The pressure in the gas bottle must always be considerably higher than the maximum working pressure of the autoclave. In order to produce the equalisation pressure compressed air could, of course also be used. If, however, an oil or other organic liquid, for example diphenyl, or a derivative thereof (Dow therm) is used as heating medium there would be a certain danger of fire in the event of a leak in the coil. If, on the other hand steam is used as heating medium there would be no objection to using compressed air. When the internal pressure increases two diaphragms 11 and 12 of the regulator are forced downwards, so that a valve 13 is opened and the gas flows from the pressure bottle 10 through the conduit 7 into the intermediate space 14. In this manner the equalisation pressure is increased. If, on the other hand, the interior pressure falls the equalisation pressure, which is then higher, forces the two diaphragms 11 and 12 upwards, so that the valve 15 of the regulator 9 is opened. The gas then passes into the atmosphere through an exhaust conduit 16. Two springs 17 and 18 ensure that the valves 13 and 15 are tightly closed when the internal pressure and equalisation pressure on each side of the diaphragms bear a certain ratio to one another.

In the conduit 6 there are provided a safety valve 19 and a control valve 20 and two manometers 21 and 22. The control valve 20 makes it possible to test at any time whether the conduit 6 is blocked between the reaction vessel and the connection of the valve. Furthermore, by comparing the readings of the manometers 21 and 22 it is possible also to test the conduit 6 during the opening of the control valve 20. The conduit 7 is likewise provided with a safety valve 23 in the pressure casing 1 and 2, a control valve 24 and a manometer 25. By opening the control valve 24 it can be ascertained by the odour of the issuing gas whether the gas from the reaction has penetrated into the space 14 due to a leakage. By comparing the readings of the manometers 25 and 21 the pressure regulators 9 and 8 can be adjusted for correct operation. Furthermore, the control valves 20 and 24 should be operated with caution in order not to falsify the pressure regulation or render it ineffective.

The equalisation pressure in an autoclave constructed in accordance with this invention is controlled automatically by the suitable operation of valves so that it is approximately equal to the reaction pressure.

In some cases it is of advantage to maintain the equalisation pressure by means of the control system at a somewhat higher value than the reaction pressure. In this manner, for example, the penetration of gases from the chemical process through accidental leakage into the intermediate space and consequent corrosion can be avoided.

The shaft 26 of a stirring device 27 is mounted in a stuffing box 28. An example of such a stuffing box is shown in Fig. 7. The inlet and outlet connections 30 and 31 for the heating medium are sealed within a stuffing box 29. The intermediate space 14 is advantageously filled with an insulating material. Fig. 8 shows an example of a stuffing box 29.

In Figs. 2—6, are shown various constructions of the interior reaction vessel. While in Fig. 1 the upper part 4 and lower part 3 of the reaction vessel are flanged together entirely independently of the pressure-resistant casing 1 and 2, this is not so in the construction shown in Figs. 1—6.

In the construction shown in Fig. 2 the reaction vessel consists of a lower part 53 and an upper part 54. These two parts are provided with flanges 153 and 154 which are so wide that they lie between the flanges of the two parts 51 and 52 of the pressure-resistant casing. A gas conduit 57, which supplies the equalisation pressure, is connected both to the cover 52 and the lower part 51 of the pressure-resistant casing. The connection with the lower part is advantageously through a stuffing box 59.

Fig. 3 shows a detail of the construction of Fig. 2. As shown in Fig. 3 the flanges 153 and 154 are worked on both sides so as to ensure good packing with the flanges of the casing 51 and 52. In order to reduce as far as possible the conduction of heat from the reaction vessel to the pressure-resistant casing the flanges are made as thin as possible and are supported between linings 151 and 152 of heat-insulating material. Furthermore, a coil 155 is arranged close to the flange 153.

In Fig. 4 is shown a third construction. In this construction the reaction vessel consists only of a lower part 73. This is permissible when it is not necessary to protect the upper part 72 of the external casing from attack by gas from the chemical process. In this way the construction of the stuffing box 78 is considerably simplified. The pressure conduit 76 is also advantageously connected to the cover 72, and this connection is substantially simplified in the construction of Fig. 4. The gas conduit 77 for the equalisation pressure is also connected with advantage to a stuffing box 79. The safety valve 23, control valve 24 and manometer 25 must then be connected to this part. In order to reduce the thermal stress in the cover 72, the latter may be provided on its outer surface with heat insulation material 80.

Figs. 5 and 6 show two ways of packing the reaction vessel 73 with respect to the lower part 71 of the exterior casing. As shown in Fig. 5 the flange 76 of the reaction vessel 73 is again screw-clamped between the flanges 71 and 72 of the exterior casing, the whole of the flange being as thin as possible as in the con-

struction shown in Fig. 3. As shown in Fig. 6 the reaction vessel 73 has a flange by means of which it is connected to the lower part 71 of the casing by means of screws 75 with the interposition of a heat-insulating lining 80. This construction has the advantage that when the cover 72 is removed the packing between 71 and 73 remains unaffected.

Fig. 7 shows a construction of stuffing box 28 which is used when the reaction vessel has an upper part 4. The sealing of the interior of the reaction vessel is ensured by a packing 33 and a ring 42. A packing 35 and a ring 36 seal the intermediate space 14.

Fig. 8 shows an example of a lower stuffing box 29, in which sealing is accomplished by means of a packing 37 and a ring 38. Channels 39, 40 and 41, respectively, in the stuffing box are the supply channel for the pressure equalisation conduit 57 (Fig. 2) and the supply conduit 30 and return conduit 31 for the heating medium (Fig. 1).

Fig. 9 shows a further form of the invention, in which the reaction vessel 103 has a relatively thick flange 104, which is clamped between the flanges of the cover 102 and the lower part 101 of the external casing. The flange 104 is sufficiently thick to enable conduits 106 and 107 for maintaining and regulating the pressure, and also a supply conduit 109 and an offtake conduit 108 communicating with the heating coil 110 on the outer surface of the reaction vessel, to be connected at the outer edge of the flange 104. Accordingly, no bores are necessary in the cover and lower portion of the outer casing, apart from the bush 128 required to accommodate the shaft of the stirring device 127. The construction shown in Fig. 9 is very advantageous, because the pressure-resistant outer casing of the autoclave is not only free from thermal stresses arising from the heating or cooling of the contents of the reaction vessel, but all bores, passages etc. which could impair the strength of the casing are dispensed with.

The construction of Fig. 9 may, of course, be modified by the provision of a reaction vessel consisting of an upper portion as well as a lower portion. In this case the upper portion of the reaction vessel will be separately fixed on the flange 104, and a passage for the supply of compressed gas to the space between the upper portion of the reaction vessel and the upper portion of the outer casing will also be provided.

An autoclave constructed in accordance with this invention has, *inter alia,* the advantage that the construction and dimensions of the pressure resistant outer casing can be chosen solely with regard to the working pressure it has to withstand and the desired size of the autoclave, and that no regard need be paid to additional stresses which would otherwise arise from the heat exchange taking place during the reaction. Thus, processes operated at high heat throughputs can be carried out without injury to the pressure-resistant outer casing, that is to say, processes in which the temperature in the reaction zone changes very rapidly. Thus, for example, the reaction vessel can be raised to the reaction temperature in a shorter time, so that, for example, the yield obtained when sensitive substances are used is substantially improved. A further advantage is that owing to the low thermal inertia of the autoclave the rates at which temperature changes are made in carrying out processes on a large scale can be made to conform to a considerable extent to the rates used in carrying out the process on the laboratory scale. The low thermal inertia of the autoclave also enables the unproductive periods of heating up and cooling between individual charges to be considerably shortened, so that the autoclave is utilised with greater economy and the cost of the process is reduced.

It will be understood that the invention is not limited to the constructions given by way of example. These constructions can of course, be modified to suit various conditions, but it is, of course, always essential to observe the basic principal, namely the carrying out of processes under high pressures and at high time-temperature and/or space-temperature gradients, that is to say processes in which large quantities of heat are supplied or abstracted in a short time, or in which the material must be heated or cooled as rapidly as possible to initiate the reaction. In accordance with the invention therefore a process is carried out in a relatively thin-walled inner reaction vessel, while the working pressure is withstood by an outer pressure-resistant casing which encloses the reaction vessel, the means for supplying or abstracting heat being directly associated with the reaction vessel itself, and the reaction vessel being heat-insulated with respect to the pressure-resistant outer casing.

WHAT WE CLAIM IS:—

1. An autoclave for carrying out processes operated at high throughputs of heat, which comprises an outer casing capable of withstanding the full working pressure within the autoclave, an inner reaction vessel having a relatively thin wall, heat exchange means forming part of, or in thermal contact with the wall of the reaction vessel for supplying heat energy to or withdrawing it from the said vessel, heat-insulating means interposed between the reaction vessel and the pressure-resistant outer casing, and means for maintaining in the space between the reaction vessel and the outer casing a pressure approximately equal to the pressure in the reaction vessel.

2. An autoclave as claimed in claim 1, wherein the heat transfer means consists of a coiled passage for a heating or cooling medium in thermal contact with the wall of the reaction vessel.

3. An autoclave as claimed in claim 2,

wherein the coiled passage is pressure-resistant so that the heating or cooling medium can be passed therethrough under atmospheric pressure.

4. An autoclave as claimed in claim 1, 2 or 3, wherein heat-insulating means takes the form of a heat-insulating material accommodated within the space between the reaction vessel and the pressure-resistant outer casing.

5. An autoclave as claimed in claim 4, wherein the heat-insulating material is permeable to gases so as to offer substantially no resistance to pressure equalisation within the said space.

6. An autoclave as claimed in claim 1, 2 or 3, wherein the heat-insulating means takes the form of a heat-insulating fluid within a space between the reaction vessel and the pressure-resistant outer casing.

7. An autoclave as claimed in any one of claims 1—5, wherein the space, within which the heat-insulating means is provided between the reaction vessel and the pressure-resistant outer casing, is connected to a source of gas under pressure through an automatic pressure equalisation device for maintaining in the said space a pressure approximately equal to the pressure within the reaction vessel.

8. An autoclave as claimed in claim 7, wherein there is provided in the said space a heat-insulating material which occupies a large proportion of the volume of the said space so that the volume of gas required for pressure equalisation is small.

9. An autoclave as claimed in claim 7 or 8, wherein the pressure-resistant outer casing is formed of a lower portion and an upper portion each having a flange opposing a flange on the other portion, and the reaction vessel is provided with a thick flange which is clamped between the opposing flanges of the lower and upper portions of the outer casing and in which are provided passages for permitting pressure equalisation between the interior of the reaction vessel and the space containing the heating-insulating means and inlet and outlet passages to enable a heating or cooling medium to be passed through the heat exchange means.

10. An autoclave as claimed in any one of claims 1—9 wherein the reaction vessel is sealed off on all sides from the pressure-resistant outer casing.

11. An autoclave as claimed in any one of claims 1—9, wherein the pressure-resistant outer casing is formed of a lower portion and an upper portion, and the reaction vessel consists only of a lower portion which is housed within the lower portion of the outer casing so as to form a gas-tight space between the latter and the reaction vessel.

12. An autoclave constructed substantially as described with reference to and shown in the accompanying drawings.

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871752 COMPLETE SPECIFICATION

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Sheet 1

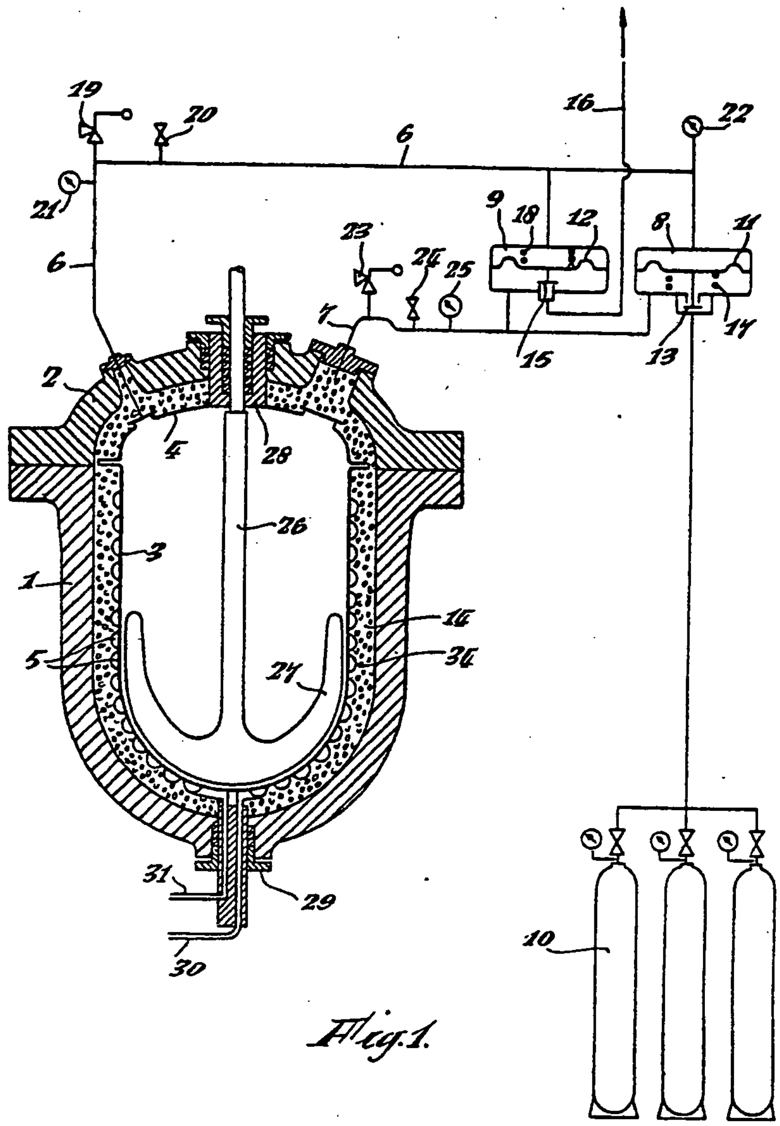

Fig.1.

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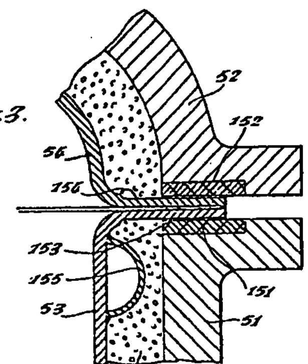


Fig.3.

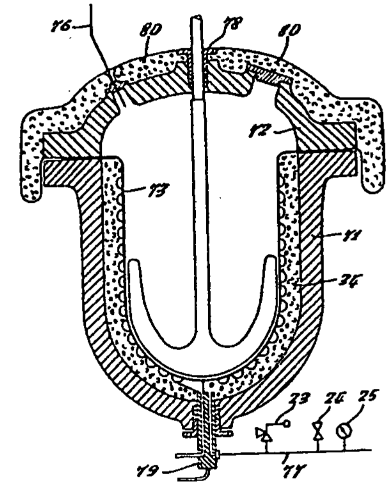


Fig.4.

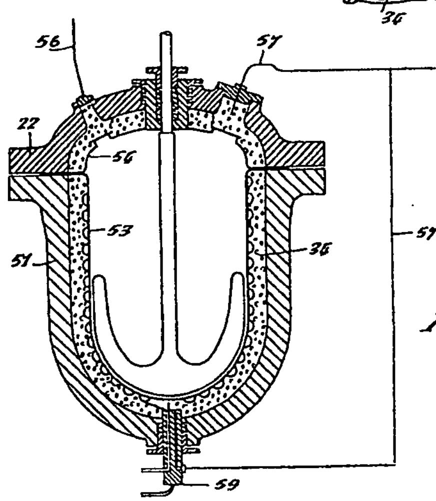


Fig.2.

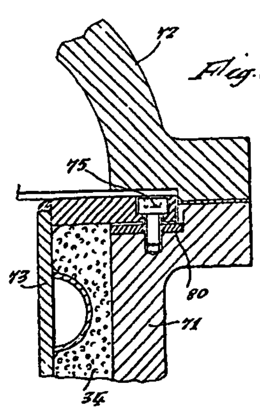


Fig.6.

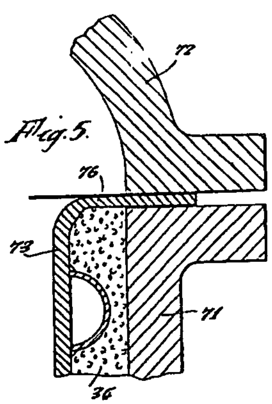
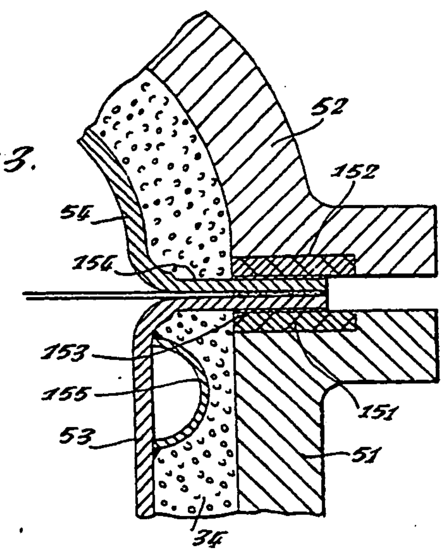


Fig.5.

Fig.3.



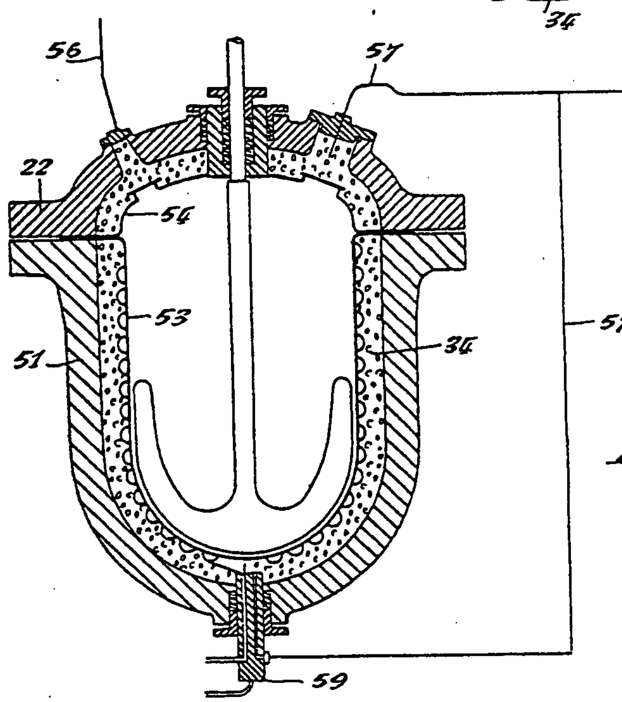


Fig.2.

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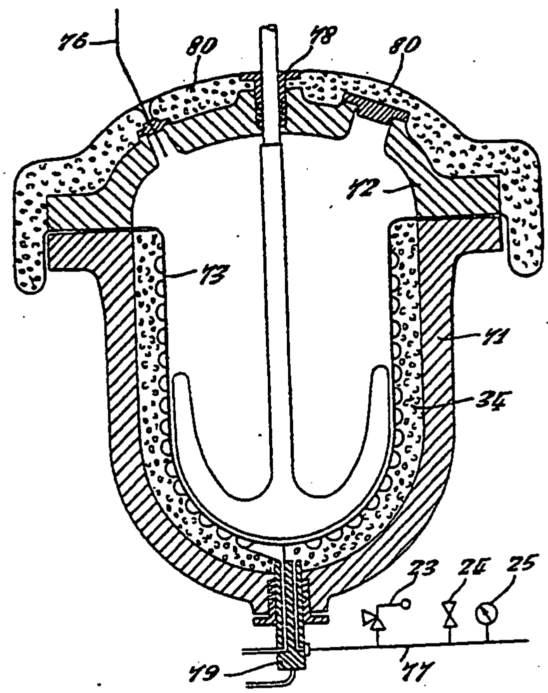
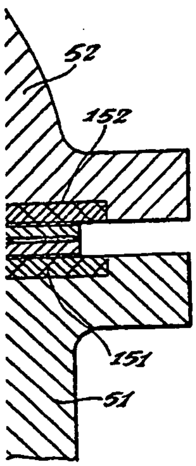
Fig.4.





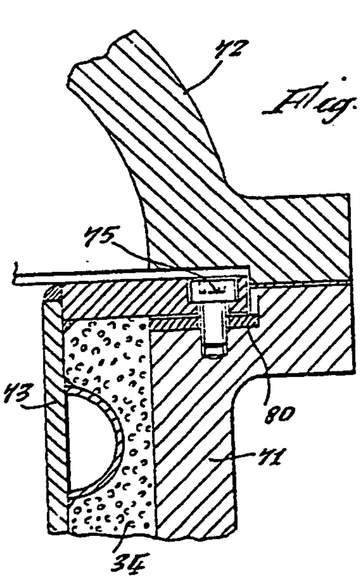


Fig.6.

Fig.5.

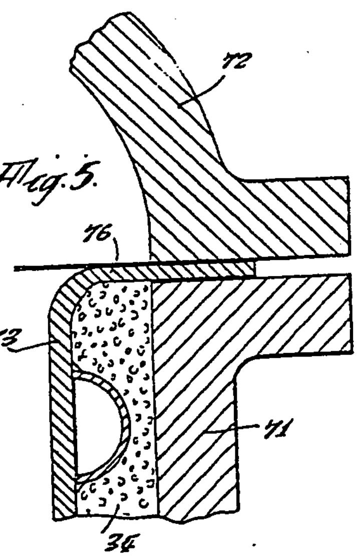


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Fig.2.

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Fig. 7.

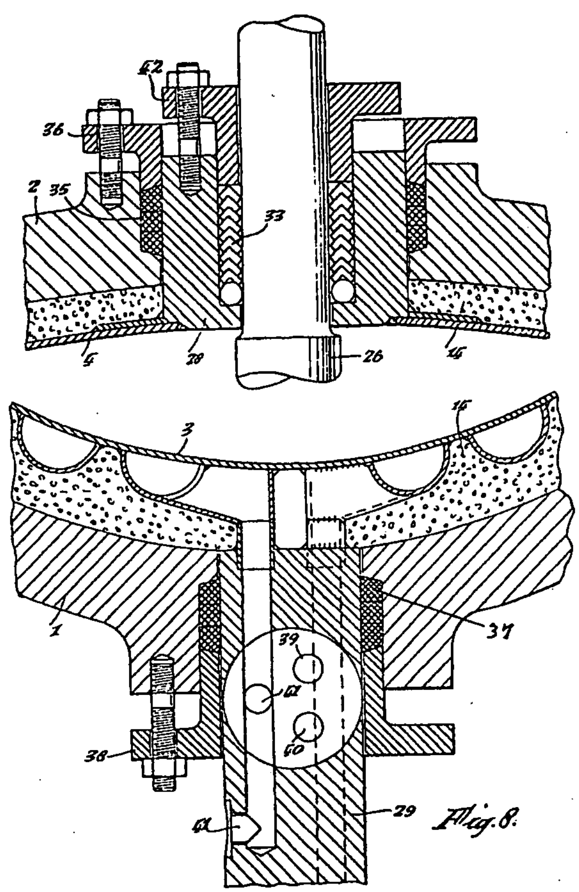


Fig. 8.

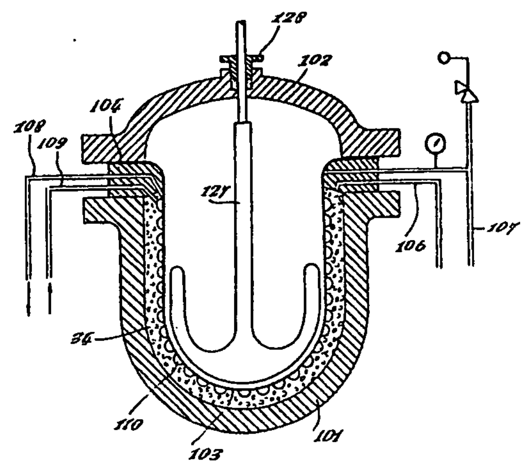
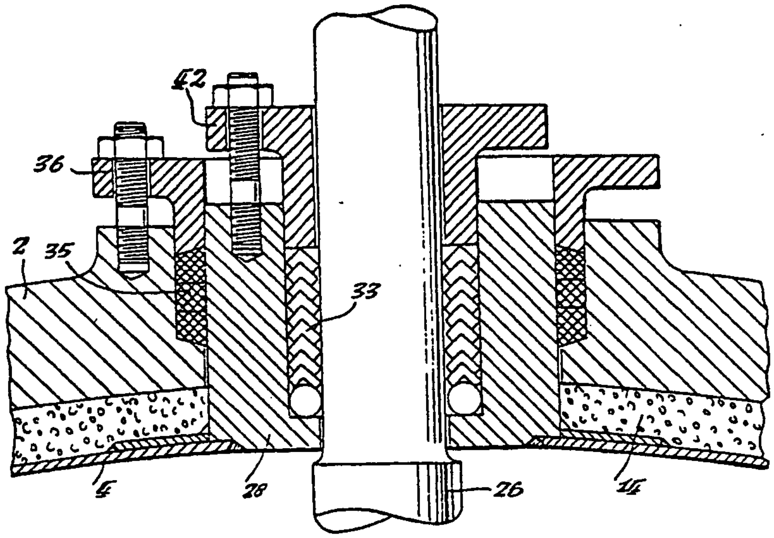


Fig. 9.

Fig. 7.



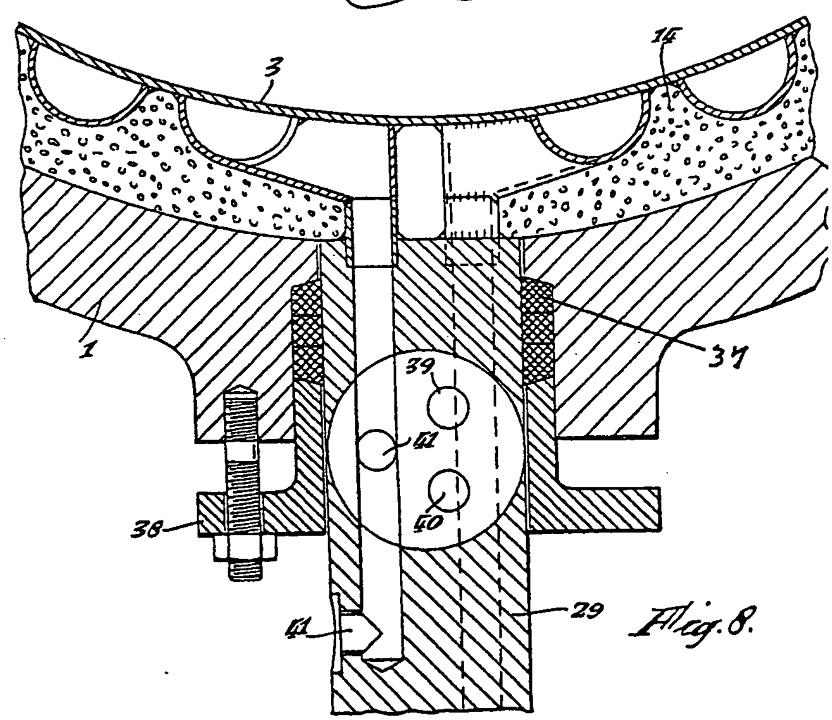


Fig. 8.

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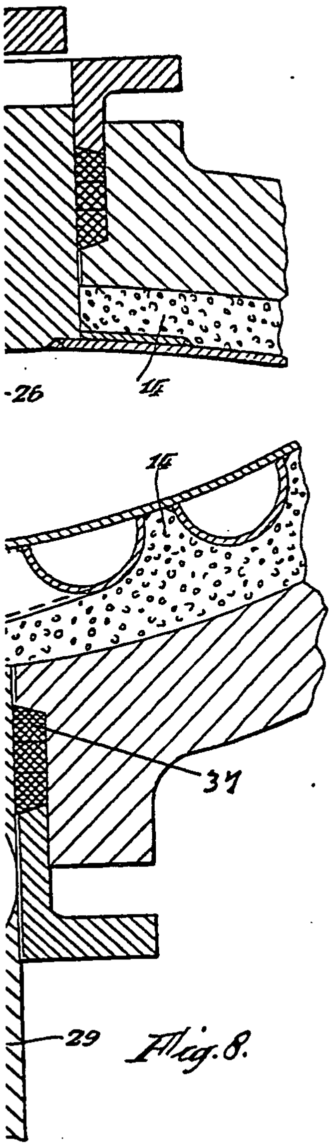

Fig. 8.

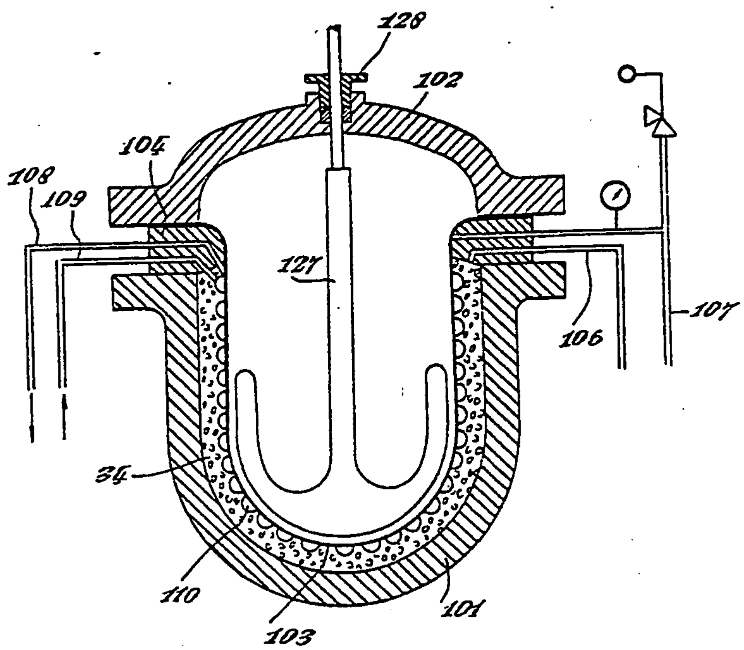

Fig. 9.